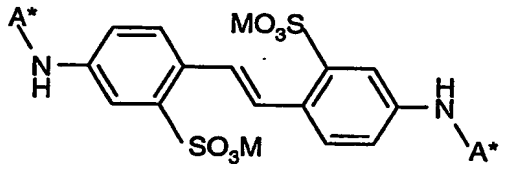
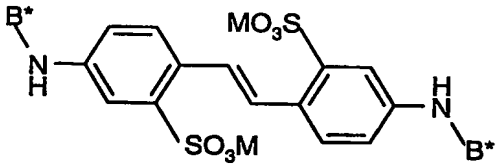
Claims

1. A fluorescent whitening agent, which comprises a mixture of compounds of the formulae

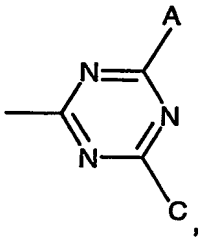

(1a),


(1b) and


(1c),

in which

A* represents a group of the formula

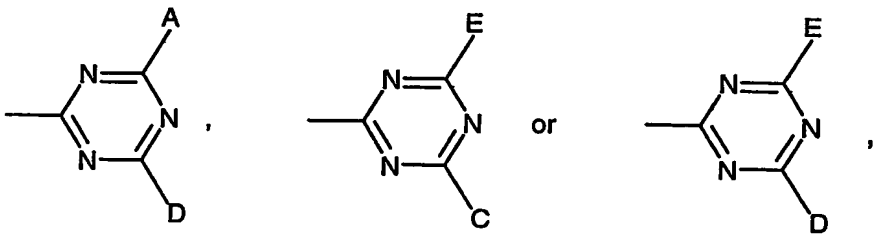


wherein

A represents –X-Y-NR_3R_4 and

C is -NR_1R_2 and

B* represents a group of the formula



wherein

D represents $-NR_5R_6$ and

E represents $-X_1-Y_1-NR_7R_8$, whereby

X and X_1 each, independently of each other, represent -O- or -NH-,

Y and Y_1 each, independently of each other, represent a straight-chain C_2-C_8alkylene or branched C_3-C_8alkylene chain, which may be interrupted by one or two nitrogen, oxygen or sulphur atoms or represent a 5- or 6-membered cycloaliphatic ring,

R_1, R_2, R_5 and R_6 each independently of each other, represent hydrogen, C_1-C_8alkyl, C_2-C_4hydroxyalkyl, C_1-C_4alkoxyC_1-C_4alkyl, phenyl, which is unsubstituted or substituted by halogen, C_1-C_4alkoxy, C_1-C_4alkyl or sulphonamido, or

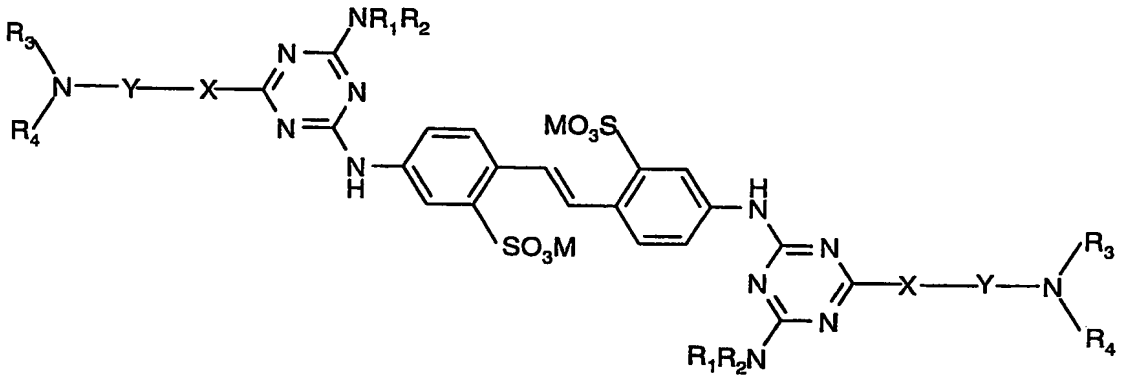
R_1 and R_2 and /or R_5 and R_6, together with the nitrogen atom to which they are attached, complete a morpholino- piperidino- or pyrrolidino-ring,

R_3, R_4, R_7 and R_8, each independently of each other, represent hydrogen, C_1-C_4alkyl, C_2-C_4hydroxyalkyl or

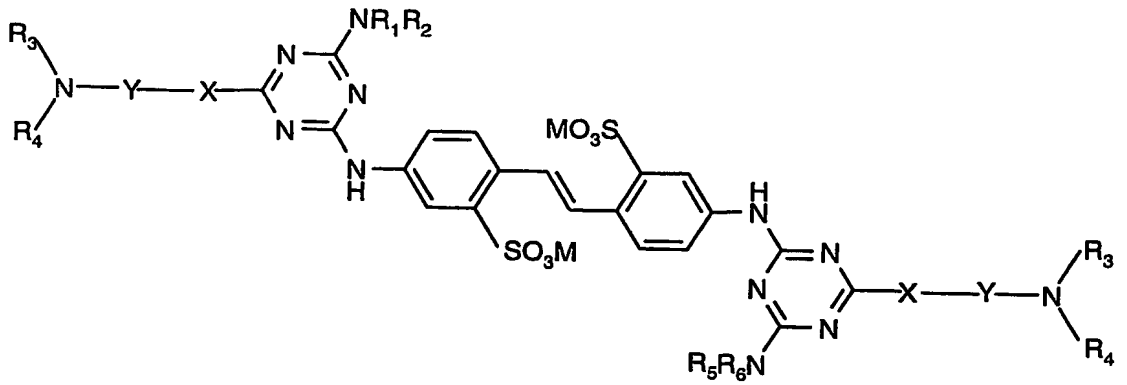
R_3 and R_4 and/or R_7 and R_8, together with the nitrogen atom to which they are attached, complete a morpholino-, piperidino- or pyrrolidino-ring and

M represents hydrogen, an alkaline or alkaline earth metal, ammonium or alkylammonium.

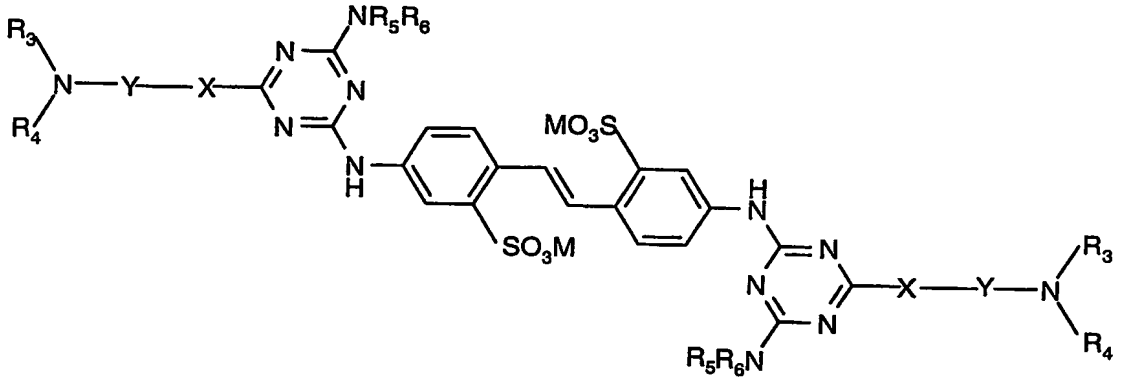
2. A fluorescent whitening agent, according to claim 1, which comprises a mixture of compounds of the formulae



(1d),



(1e) and

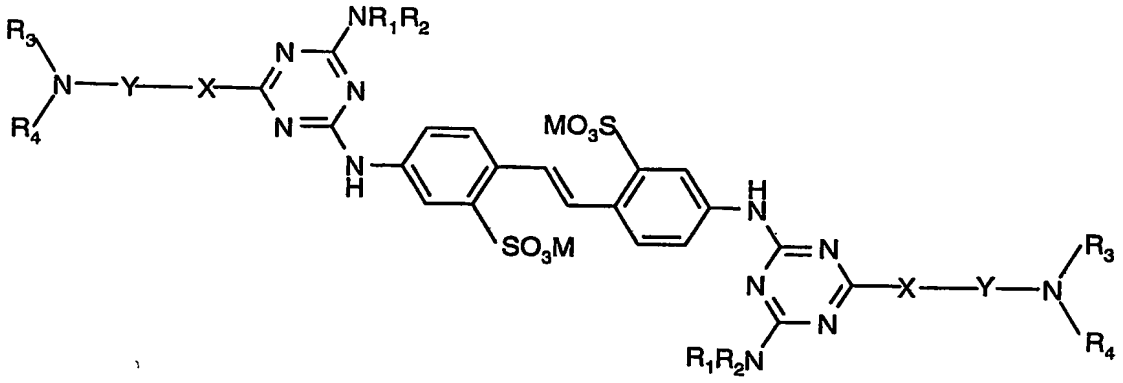


(1f),

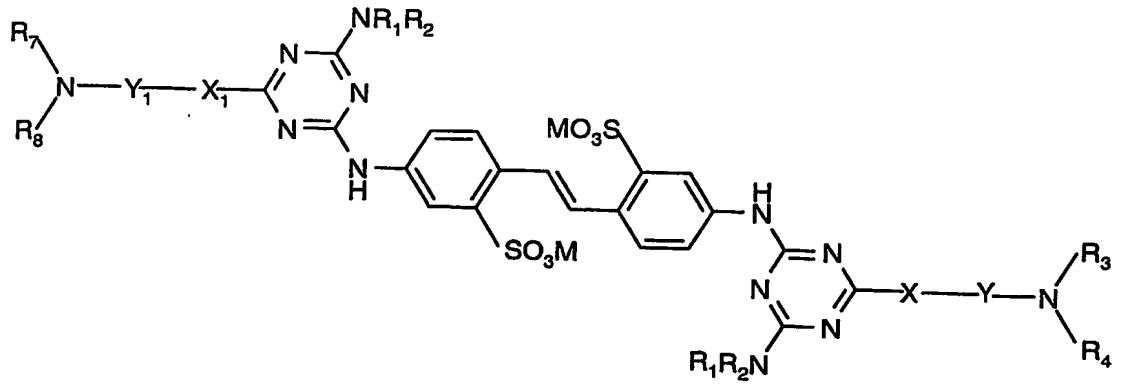
in which

X, Y, R_1, R_2, R_3, R_4, R_5, R_6 and M are as defined in claim 1.

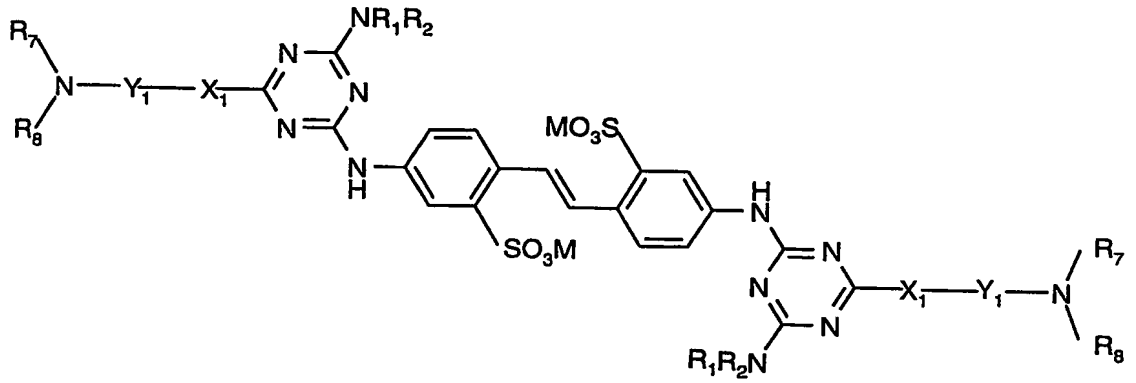
3. A fluorescent whitening agent, according to claim 1, which comprises a mixture of compounds of the formulae



(1d),



(1g) and

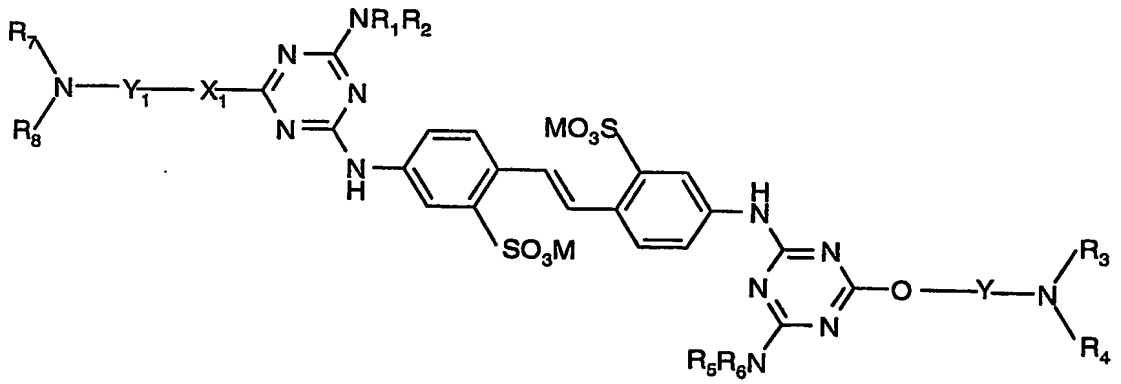


(1h),

in which

X, X_1, Y, Y_1, R_1, R_2, R_3, R_4, R_7, R_8 and M are as defined in claim 1.

4. A compound of formula

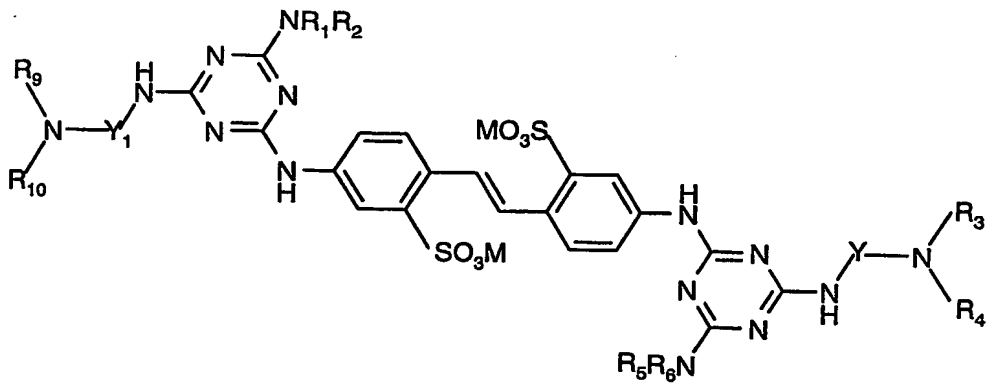


(2),

in which

X_1, Y, Y_1, R_1, R_2, R_3, R_4, R_5, R_6, R_7, R_8 and M are as defined in claim 1.

5. A compound of the formula



(3),

in which

R_9 and R_{10}, each independently of each other, represent hydrogen or C_2-C_4hydroxyalkyl and Y, Y_1, R_1, R_2, R_3, R_4, R_5, R_6, and M are as defined in claim 1, with the proviso that when Y and Y_1 both represent $-CH_2CH_2CH_2-$, R_1 and R_5 are both phenyl and R_2 and R_6 are both hydrogen, R_3, R_4, R_9 and R_{10} are not all $-CH_2CH_2OH$.

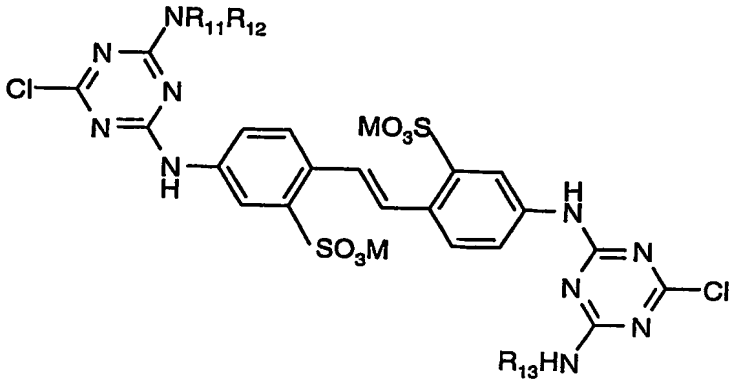
6. A process for the preparation of a mixture of compounds of formulae (1a), (1b) and (1c), according to claim 1, by reacting, under known reaction conditions, cyanuric chloride, successively, in any desired sequence, with each of 4,4'-diaminostilbene-2,2'- disulphonic acid, amino compounds of formulae R_1R_2NH and R_5R_6NH or mixtures thereof and compounds of formulae R_3R_4YXH and $R_7R_8Y_1X_1H$ or mixtures thereof,
X, X_1, Y, Y_1, R_1, R_2, R_3, R_4, R_5, R_6, R_7 and R_8 being as defined in claim 1.

7. A process for the preparation of a compound of formula (2), according to claim 4, by reacting, under known reaction conditions, cyanuric chloride, successively, in any desired sequence, with each of 4,4'-diaminostilbene-2,2'- disulphonic acid, an amino compound of formula R_1R_2NH, an amino compound of formula R_5R_6NH, a hydroxy compound of formula R_3R_4NYOH and a compound of formula $R_7R_8NY_1X_1H$,
X_1, Y, Y_1, R_1, R_2, R_3, R_4, R_5, R_6, R_7 and R_8 being as defined in claim 1.

8. A process for the preparation of a compound of formula (3), according to claim 5, by reacting, under known reaction conditions, cyanuric chloride, successively, in any desired sequence, with each of 4,4'-diaminostilbene-2,2'- disulphonic acid, an amino compound of

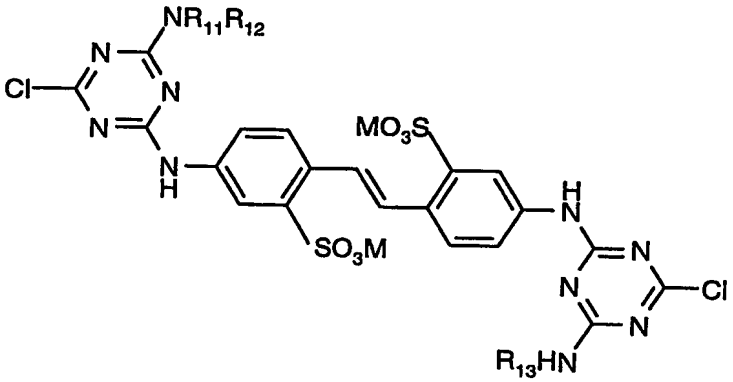
formula R_1R_2NH, an amino compound of formula R_5R_6NH, an amino compound of formula $R_3R_4NYNH_2$ and a compound of formula $R_9R_{10}NY_1NH_2$,
Y, Y_1, R_1, R_2, R_3, R_4, R_5, R_6, R_9 and R_{10} being as defined in claims 1 and 5.

9. A compound of the formula

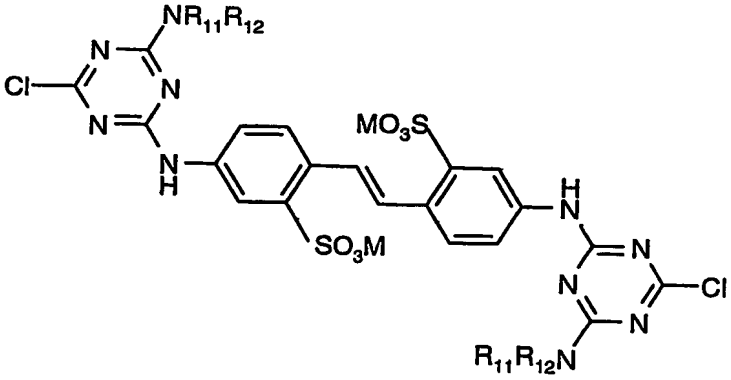


(4a)

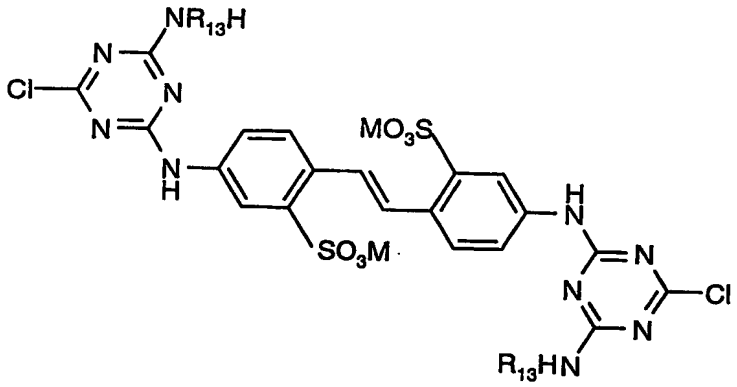
or a mixture comprising compounds of the formulae



(4a),



(4b) and



(4c),

in which

R_{11} and R_{12}, each independently of each other, represent hydrogen, C_1-C_4alkyl, C_2-C_4hydroxyalkyl, C_1-C_4alkoxyC_1-C_4alkyl or, together with the nitrogen atom to which they are attached, complete a morpholino-, piperidino- or pyrrolidino-ring,

R_{13} represents phenyl, which is unsubstituted or substituted by halogen, C_1-C_4alkoxy, C_1-C_4alkyl or sulphonamido and

M represents hydrogen, an alkaline or alkaline earth metal, ammonium or alkyl ammonium.

10. A process for the preparation of a compound of formula (4a) or a mixture of compounds of formulae (4a), (4b) and (4c), according to claim 9, by reacting, under known reaction conditions, cyanuric chloride, successively, in any desired sequence, with each of 4,4'-diaminostilbene-2,2'- disulphonic acid, an amino compound of formula $R_{11}R_{12}$ NH and an amino compound of formula $R_{13}NH_2$ or with a mixture of amino compounds $R_{11}R_{12}$NH and $R_{13}NH_2$, R_{11}, R_{12} and R_{13} being as previously defined in claim 9.

11. Use of the compound of formula (4a), according to claim 9, for the preparation of a compound of formula (2), according to claim 3, in which, in formula (2),

R_1 and R_2 each independently of each other, represent hydrogen, C_1-C_4alkyl, C_2-C_4hydroxyalkyl, C_1-C_4alkoxyC_1-C_4alkyl or, together with the nitrogen atom to which they are attached, complete a morpholino-, piperidino- or pyrrolidino-ring,

R_5 represents phenyl, which is unsubstituted or substituted by halogen, C_1-C_4alkoxy, C_1-C_4alkyl or sulphonamido,

R_6 represents hydrogen and

X_1, Y, Y_1, R_3, R_4, R_7, R_8 and M are as defined in claim 1;

for the preparation of compound of formula (3), according to claim 5, in which, in formula (3),

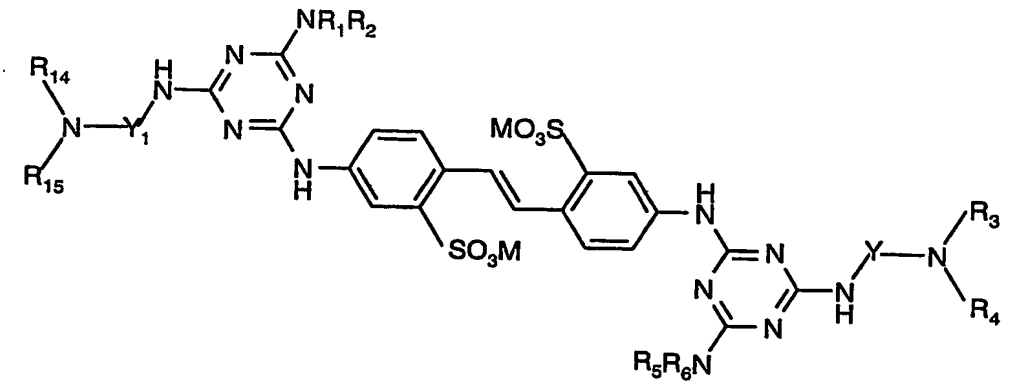
R_1 and R_2 each independently of each other, represent hydrogen, C_1-C_4alkyl,

C_2-C_4hydroxyalkyl, C_1-C_4alkoxyC_1-C_4alkyl or, together with the nitrogen atom to which they are attached, complete a morpholino-, piperidino- or pyrrolidino-ring,
R_5 represents phenyl, which is unsubstituted or substituted by halogen, C_1-C_4alkoxy, C_1-C_4alkyl or sulphonamido,
R_6 represents hydrogen and
Y, Y_1, R_3, R_4, R_9, R_{10}, and M are as previously defined in claims 1 and 5 respectively or use of the mixture of compounds of formulae (4a), (4b) and (4c), according to claim 9, for the preparation of a mixture of compounds of formulae (1a), (1b) and (1c), according to claim 1, in which, in formulae (1a), (1b) and (1c),
R_1 and R_2 each independently of each other, represent hydrogen, C_1-C_4alkyl, C_2-C_4hydroxyalkyl, C_1-C_4alkoxyC_1-C_4alkyl or, together with the nitrogen atom to which they are attached, complete a morpholino-, piperidino- or pyrrolidino-ring,
R_5 represents phenyl, which is unsubstituted or substituted by halogen, C_1-C_4alkoxy, C_1-C_4alkyl or sulphonamido,
R_6 represents hydrogen and
X, X_1, Y, Y_1, R_3, R_4, R_7, R_8 and M are as defined in claim 1.

12. Use of the mixture of compounds of formulae (1a), (1b) and (1c), according to claim 1, for the fluorescent whitening of paper.

13. Use of the compound of formula (2), according to claim 4, for the fluorescent whitening of paper.

14. Use of the compound of formula



(5),

in which

R_{14} and R_{15}, each independently of each other, represent hydrogen, C_1-C_4alkyl or C_2-C_4hydroxyalkyl and
Y, Y_1, R_1, R_2, R_3, R_4, R_5, R_6, and M are as defined in claim 1, for the fluorescent whitening of paper.

15. Paper, which has been treated with a fluorescent whitening agent comprising either a mixture of compounds of formulae (1a), (1b) and (1c), according to claim 1, a compound of formula (2), according to claim 4 or a compound of formula (5), according to claim 14.